XPCS studies of slow, non-diffusive dynamics in glassy soft matter

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For many disordered soft solids -- such as foams, gels, concentrated emulsions, and dense colloidal suspensions -- the solid phase is separated from a fluid state by an ergodic-nonergodic transition that leaves the system in an out-of-equilibrium configuration. Often such out-of-equilibrium materials display a protracted evolution of their dynamical properties that bears strong resemblance to the phenomenon of aging seen in molecular glasses and glassy polymers. Experiments on aging have typically measured the temporal evolution of response functions, such as the elastic moduli, and far fewer studies have characterized dynamical correlation functions in aging systems. In an effort to understand the microscopic dynamical behavior of out-of-equilibrium disordered materials, we have conducted x-ray photon correlation spectroscopy (XPCS) studies on a series of disordered soft solids. The combination of large wave vectors and long time scales accessible with XPCS makes the technique uniquely well suited for elucidating the nanoscale motions in such glassy materials.

The systems we have investigated include suspensions of the synthetic clay laponite [1], which slowly evolve from fluid to solid in a manner akin to aging, as well as concentrated nanoemulsions [2] and depletion gels [3], whose dynamical evolution we tracked after the cessation of a strong, fluidizing shear. In the studies involving shear, our motivation was to compare the dynamical recovery of a jammed system following shear flow with aging in glasses, which jam through a quench in temperature. For all the disordered soft solids, we observed the same generic slow dynamics characterized by an intermediate scattering function that follows a "compressed" exponential form, $g_1(q,t) \sim \exp[-(t/\tau)^{\beta}]$, with $\beta \approx 1.5$ and $\tau \sim q^{-1}$. Such behavior contrasts with glassy diffusion, for which correlation functions are stretched ($\beta < 1$) and $\tau \sim q^{-2}$. Thus, we conclude that the dynamical evolution of the disordered soft solids, while displaying signatures of aging, cannot be directly related to traditional aging phenomena in glasses.

Instead, these results indicate a type of slow, non-diffusive dynamics that are apparently universal to a range of disordered soft materials. In particular, our observations can be understood in the context of a recent model that describes the dynamics in disordered elastic media in terms of strain from random, highly localized stress relaxation events. Within this picture, the constituents of the jammed systems move with a broad distribution of strain velocities. This talk will provide an overview of these studies with an emphasis on recent work to uncover the microscopic origins of this non-diffusive motion. In particular, insights gleaned from further examination of the XPCS results, including analysis of the instantaneous two-time correlation functions for these evolving systems, will be discussed.

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[3] B. Chung, S. Ramakrishnan, R. Bandyopadhyay, D. Liang, C. F. Zukoski, J. L. Harden, and R. L. Leheny, "Microscopic Dynamics of Recovery in Sheared Depletion Gels," Phys. Rev. Lett. **96**, 228301 (2006).